Sensitivity of tropospheric chemistry simulations to cloud vertical distribution and optical properties

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Objectives

- To **improve** our understanding of the radiative effect of clouds on global trop chem
- To **quantify** the radiative effect of clouds on photolysis rates and key oxidants

    1). impact of cloud overlap assumptions

    2). sensitivity to cloud optical properties

Outline

- Objectives & GEOS-CHEM (Fast-J)
- GEOS-3 Cloud Distributions and Evaluation
- Radiative Effect on J-values and Oxidants
- Sensitivity to Cloud Vertical Distribution
  - GEOS1-STRAT → GEOS-3 → GEOS-4
- Sensitivity to Cloud Optical Depth Magnitude
  - COD adjusted progressively: 0 – 200%
- Sensitivity to Cloud Absorption: A Cautionary Note
- Conclusions

GEOS-CHEM Global Chemical Transport Model (http://www-as.harvard.edu/chemistry/trop/geos)

- Driven by GEOS1-STRAT, GEOS-3, and GEOS-4 from GMAO, $4^o x 5^o$
- Ozone-NO_x-CO-VOC coupled to aerosol (sulfate-nitrate-ammonium and carbonaceous) chemistry [Bey et al., 2001; Park et al., 2004]
- Photolysis rate calculation: **Fast-J** [Wild et al., 2000] with 3-D cloud optical depth and cloud fraction taken from GEOS
- Sensitivity simulation: 1996; Aug 2000 – Dec 2001

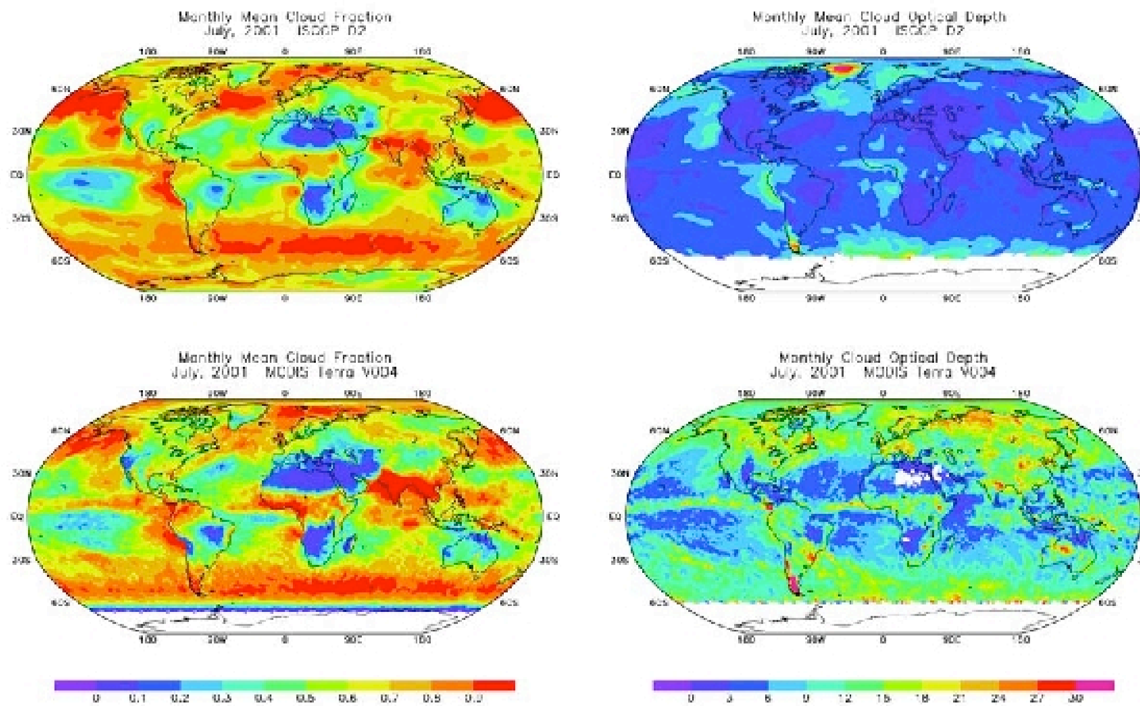
Are the MODIS and ISCCP cloud optical depths really so different? (NO)

Cloud Fraction
July, 2001

Cloud OD
July, 2001

ISCCP

MODIS



Global Energy and Water Cycle Experiment (GEWEX) NEWS,
Vol. 13, No.4, Nov 2003

ISCCP Definitions of Cloud Optical Thicknesses and Water Paths

- **Optical thickness** values from individual pixels are averaged with **non-linear** weights that preserve the average cloud albedo.

- **Water path** values are stored in the D1 and D2 datasets as optical thickness values, but they present **linear** averages of individual pixel values of optical thickness proportional to cloud water content.

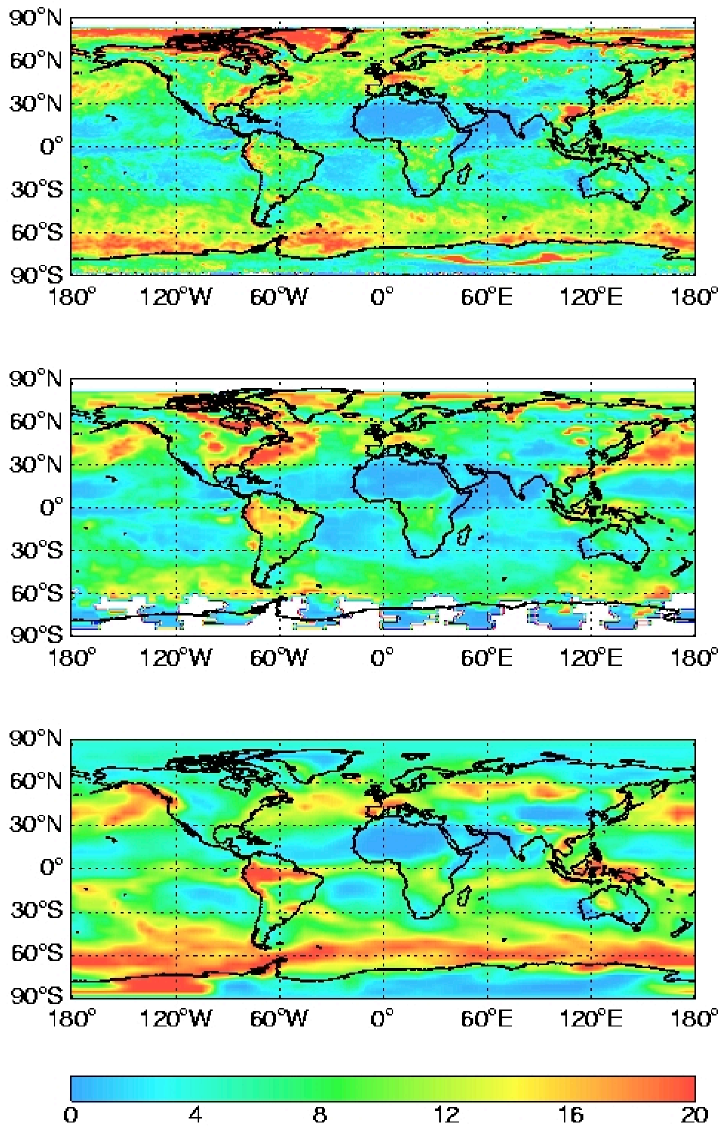
Rossow et al. [1996]

Global Distribution of Cloud Optical Depth
GEOS-3 vs. Satellite Retrievals

MODIS (MOD08_M3)

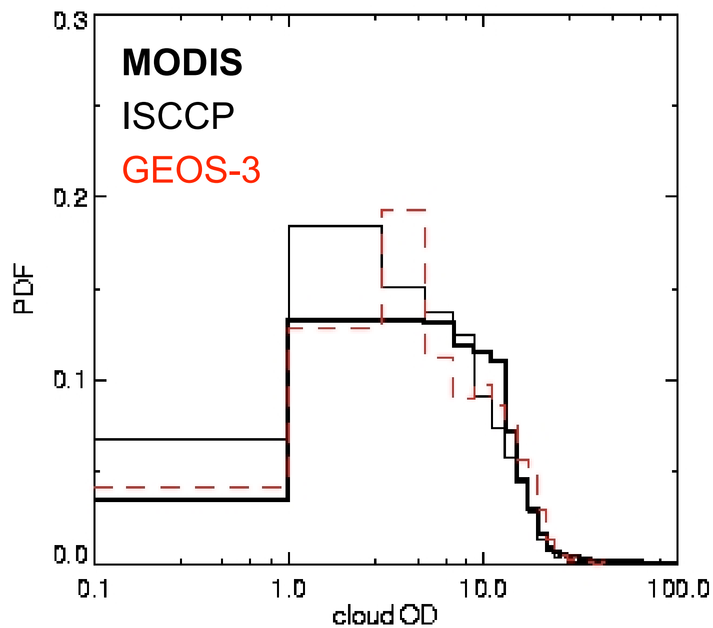
ISCCP (D2)

GEOS-3



Mean Cloud Optical Depth (grid-scale) for March 2001

Probability Distribution Functions

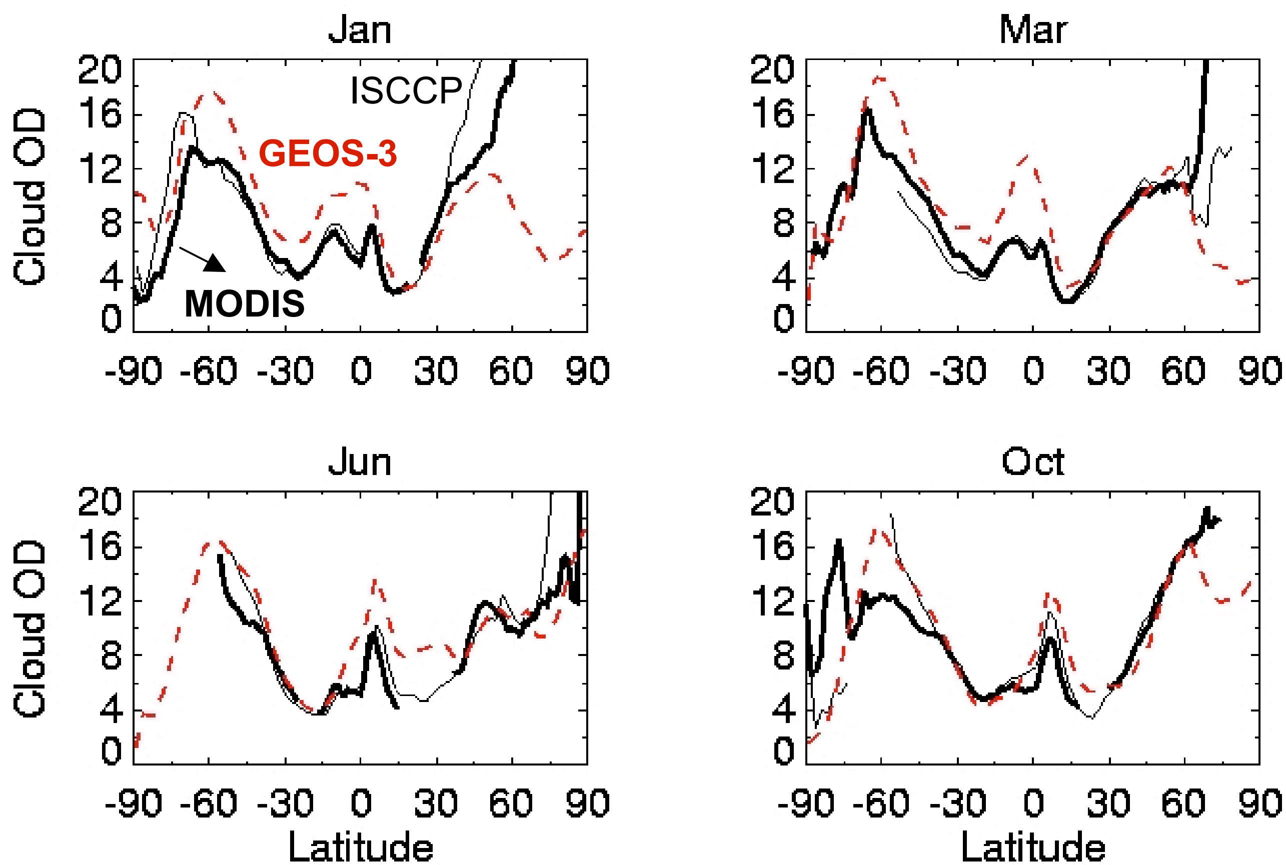


Global Average

GEOS3-OD / MODIS-OD = 0.91

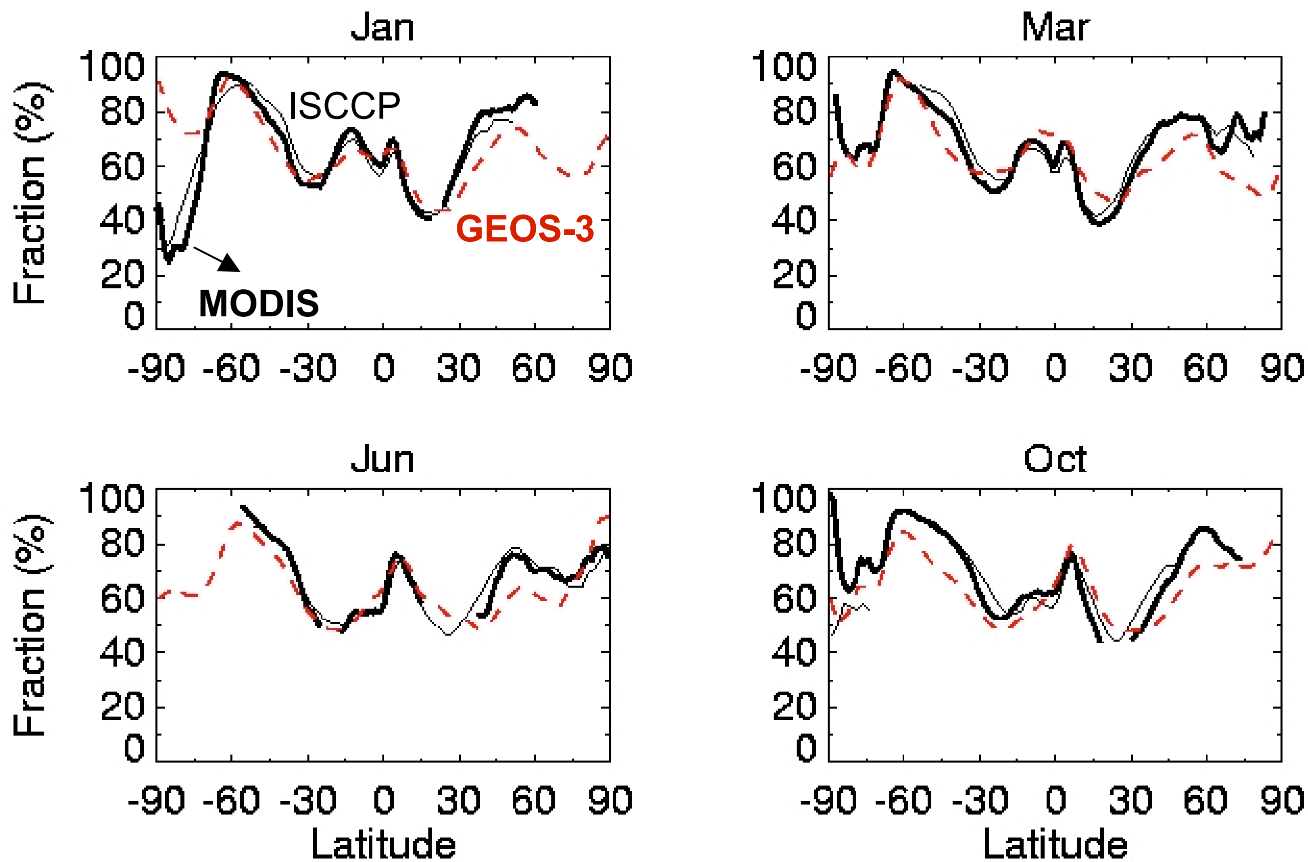
GEOS3-OD / ISCCP-OD = 1.31

Latitudinal Distribution of Cloud Optical Depth
GEOS-3 vs. Satellite Retrievals



GEOS-3 cloud OD reasonably agree with MODIS and ISCCP cloud retrieval products, but tend to be larger in the tropics and SH marine stratiform clouds region.

Cloud Fraction, 2001
GEOS-3 vs. Satellite Retrievals



GEOS-3 cloud fraction overall agrees with MODIS and ISCCP products, but tends to be lower at mid-latitudes.

Model Representations of the Vertical Coherence of Clouds

- **LIN: Linear Assumption [Wild et al., 2000]**

$$\tau_c' = \tau_c \cdot f$$

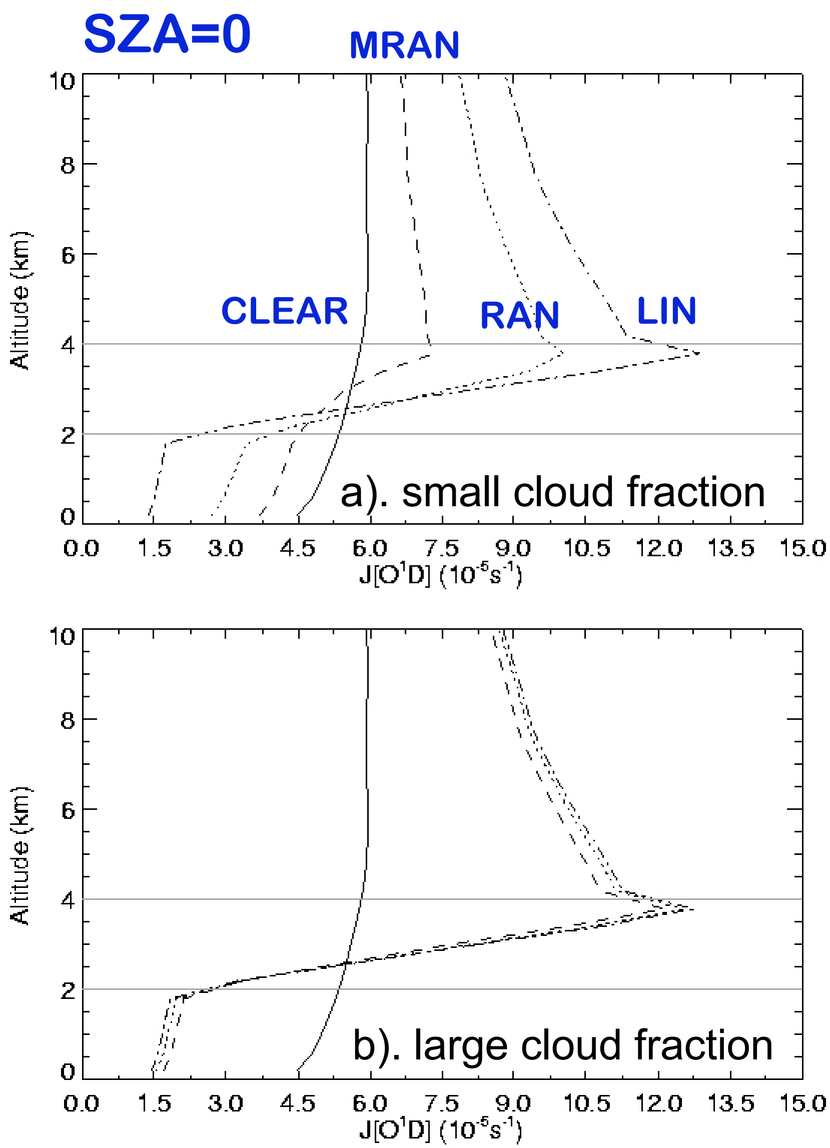
grid-scale OD   in-cloud OD   cloud fraction

- **RAN: Approximate Random Overlap [Briegleb, 1992]**

$$\tau_c' = \tau_c \cdot f^{3/2}$$

- **MRAN: Maximum-Random Overlap**
  - clouds in adjacent layers (a cloud block) are maximally overlapped; cloud blocks are randomly overlapped.
  - version 1: Stubenrauch et al. [1997];Tie et al. [2003]
  - version 2: Collins [2001]; Feng et al. [2004]

Effect of clouds on J[O^1D] calculated by off-line Fast-J (Test case of Feng et al. [2004])



Cloud layers at

3-4km: f = 0.2, **0.9**

2-3km: f = 0.1, **0.8**

Latitude = 45N

Albedo=0.1

Mean OD = 54

Enhancement above cloud

Reduction below cloud

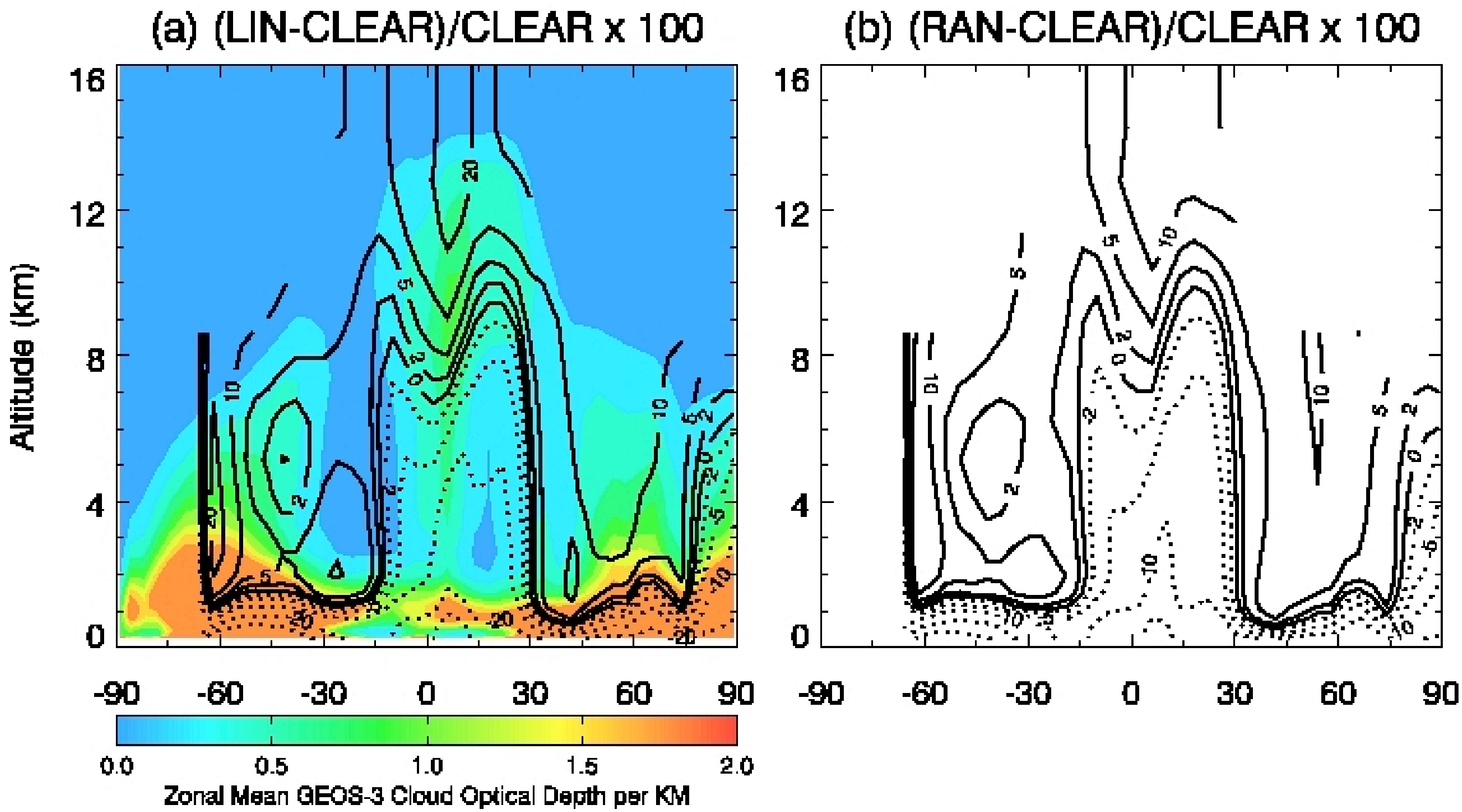
a). Small cloud fraction:

LIN > RAN > MRAN

b). Large cloud fraction:

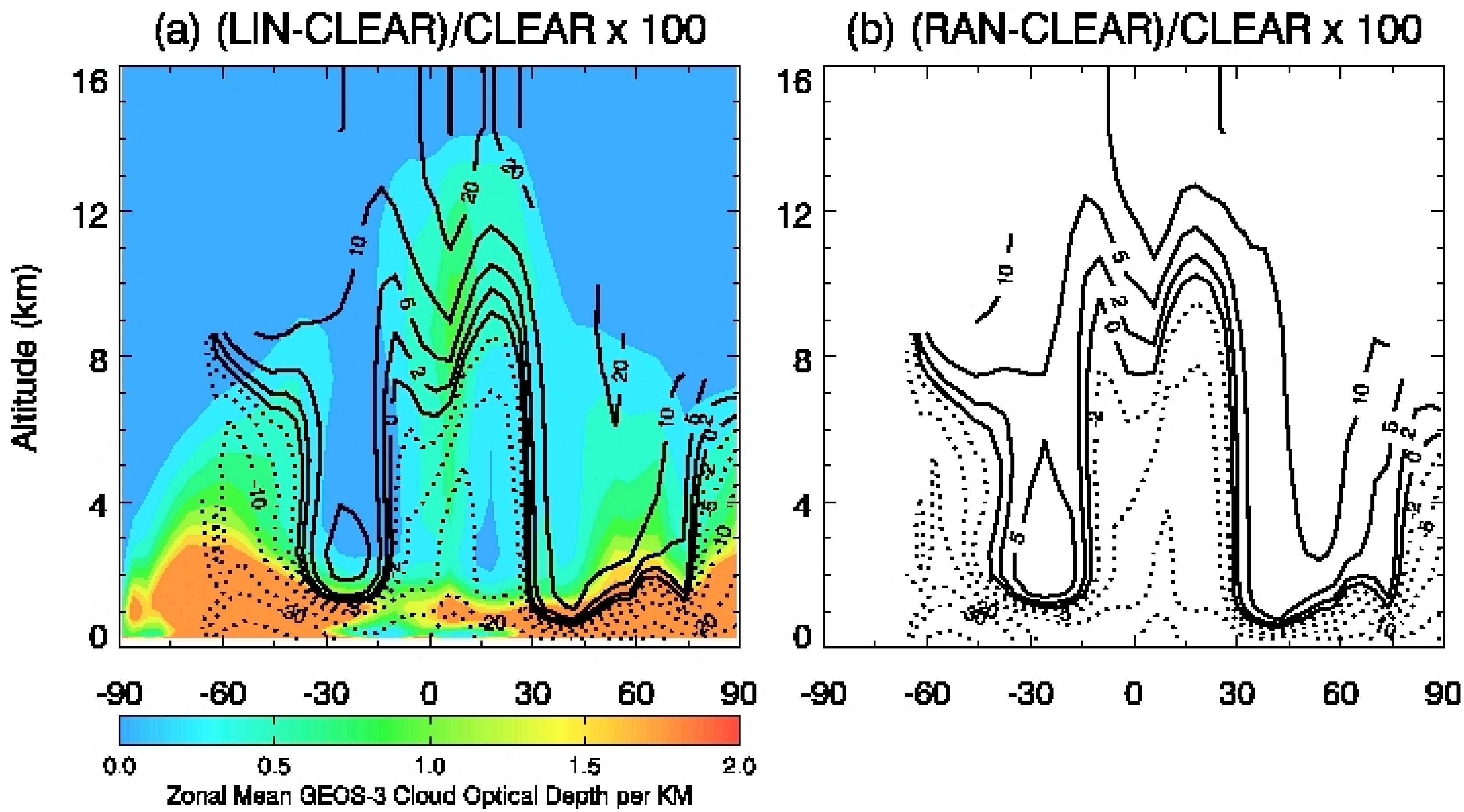
Small differences between schemes

Changes (%) in daily mean J[O^1D] due to cloud
June 2001



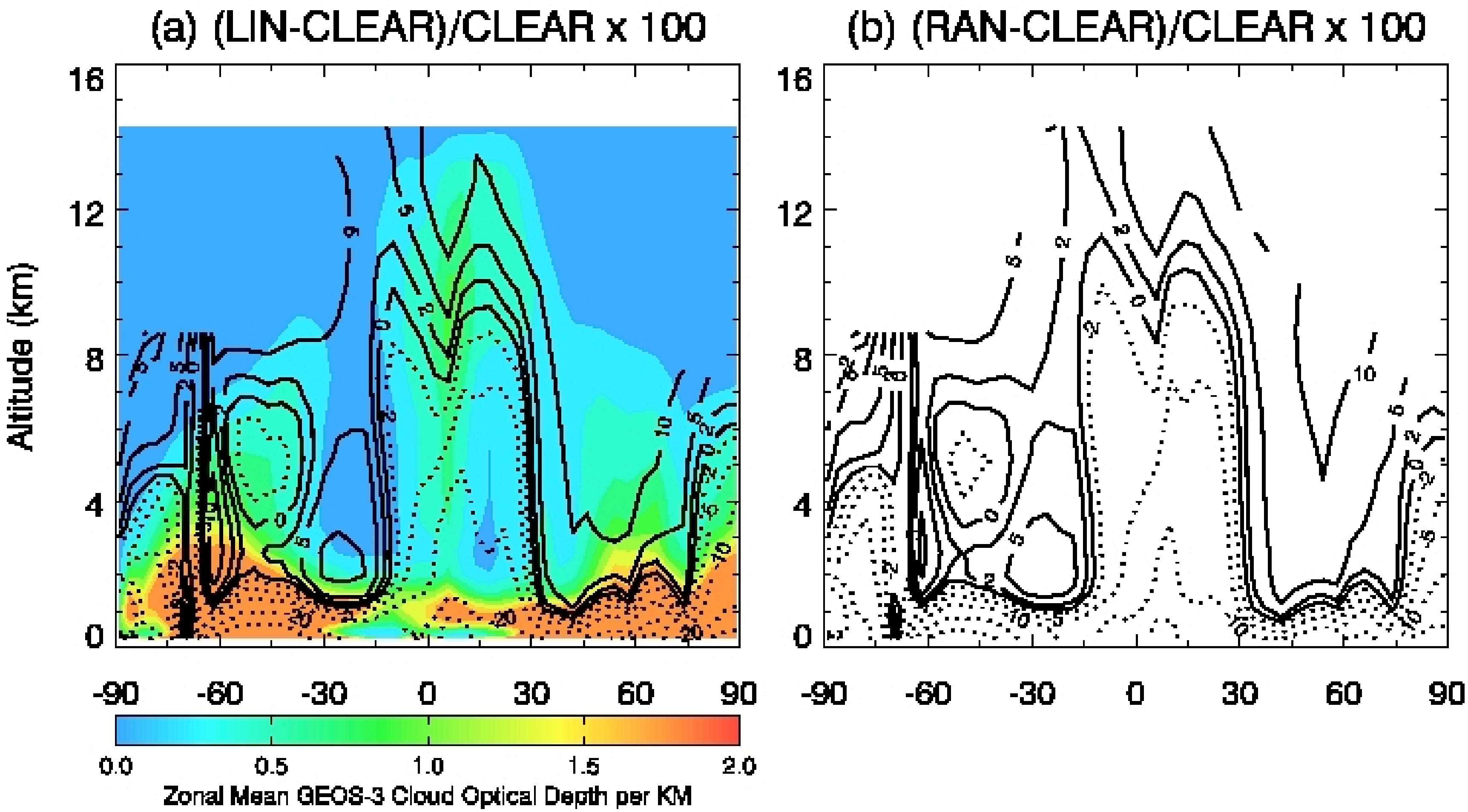
- RAN and MRAN differs by ~2%

Changes (%) in daily mean J[NO_2] due to cloud
June 2001



- RAN and MRAN differs by ~2%

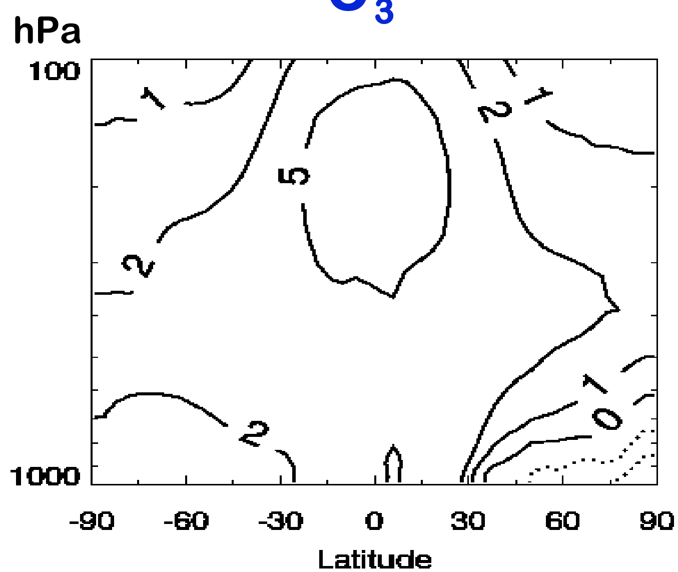
Changes (%) in daily mean OH due to cloud June 2001

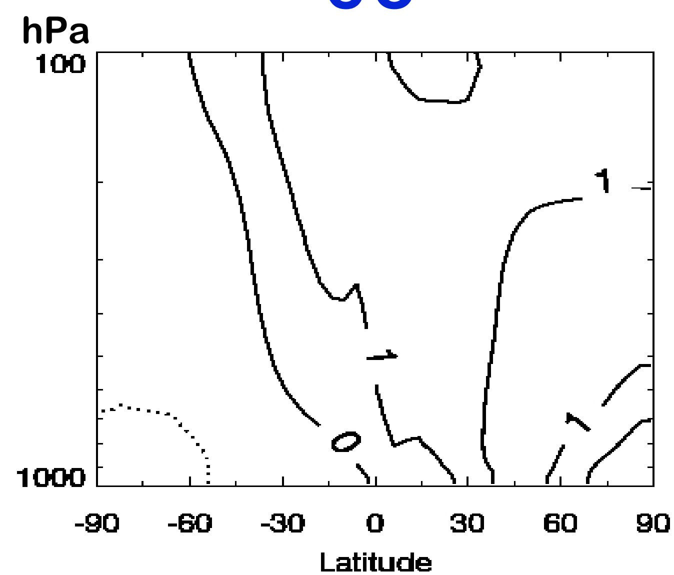


O_3 and CO changes (%) due to cloud, June (GEOS-CHEM vs. MOZART-2)

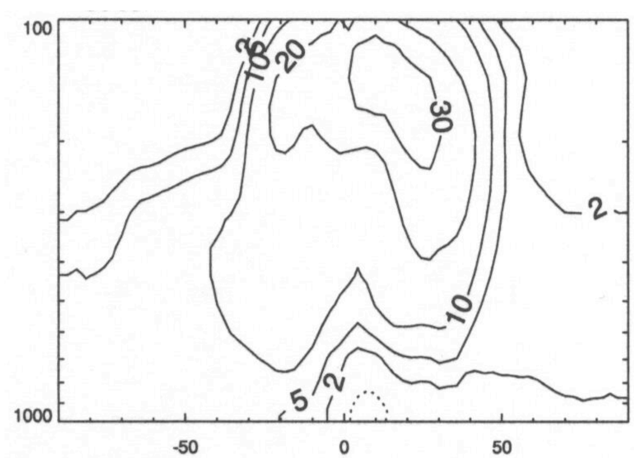
O_3 | CO

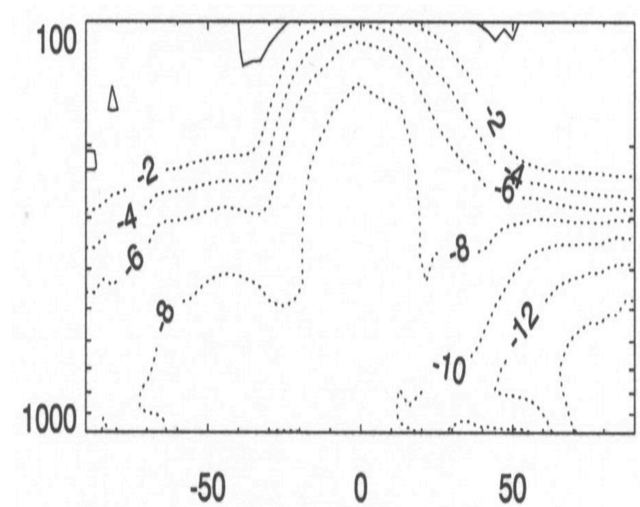
GEOS-CHEM
MRAN
[this work]





MOZART-2
MRAN
[Tie et al., 2003]



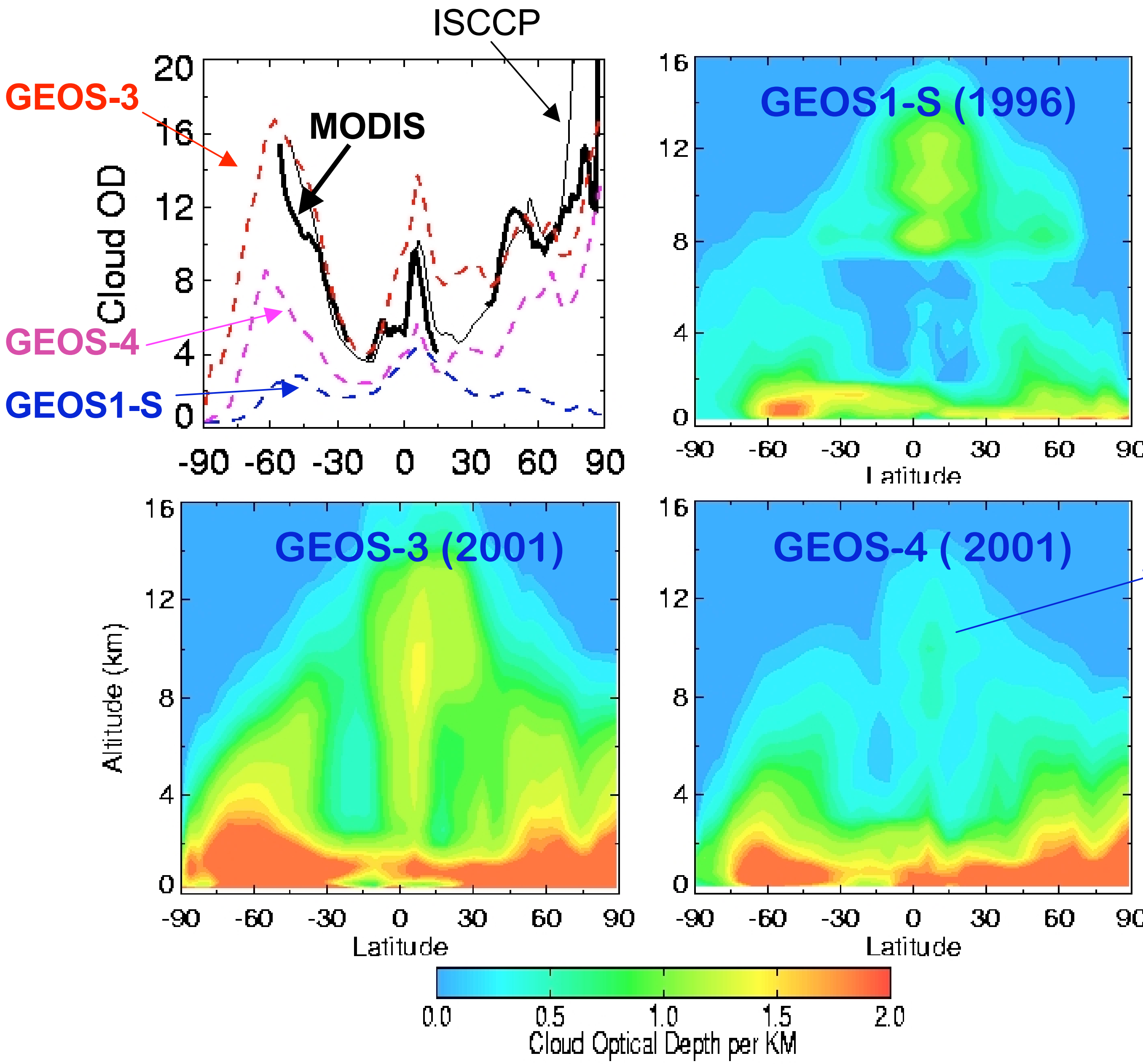


Why are the sensitivities to cloud so different ?

Global (troposphere) mean changes (%) due to cloud, June

	GEOS-CHEM [this work]			MOZART-2 [Tie et al.,2003]	
	LIN	RAN	MRAN	LIN	MRAN
OH	0.99	0.13	-0.52	88.09	20.31
O_3	4.89	3.15	3.65	12.07	8.55
NO_x	5.58	3.46	3.26	-4.17	-3.13
HO_2	-2.27	-1.60	-1.47	16.52	5.89
CH_2O	5.55	3.85	4.77	-14.56	-5.78
CO	0.81	1.33	2.26	-31.40	-9.01
$J[O^1D]$	-3.30	-2.15	-3.44	44.98	13.38
$J[NO_2]$	-4.38	-3.23	-3.76	62.24	13.84
$J[CH_2O]$	-2.30	-1.74	-2.38	54.56	13.75

GEOS1-STRAT, GEOS-3 and GEOS-4 Cloud Optical Depths per KM (June)



Global Average:

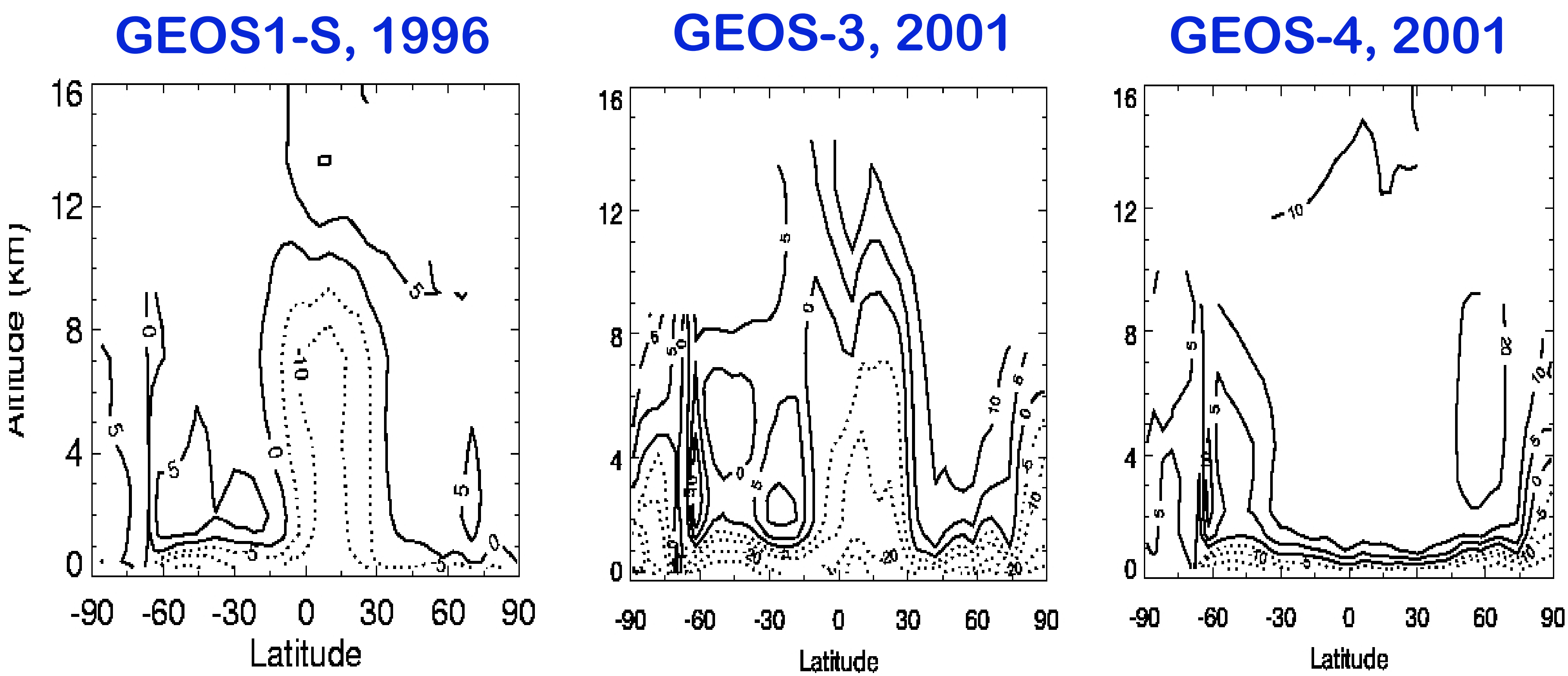
GEOS-3 / GEOS1-S = 5.1

GEOS-3 / GEOS-4 = 1.9

GEOS1-S COD: too small

optically too thin

Changes (%) in daily mean OH due to cloud (LIN, June)

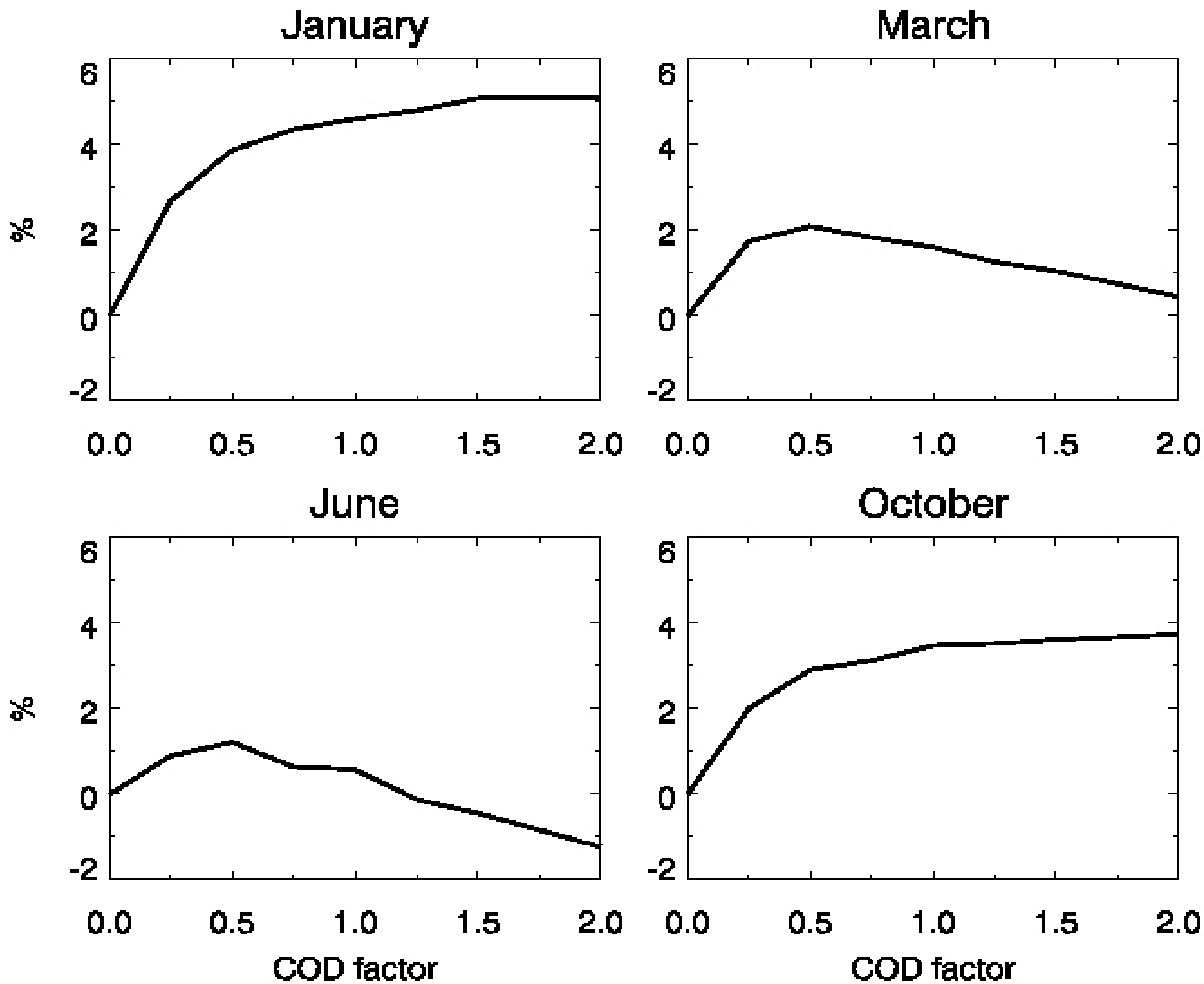


changes of global mean OH due to cloud

GEOS1-S: -1% **GEOS-3: 1%** **GEOS-4: 14%**

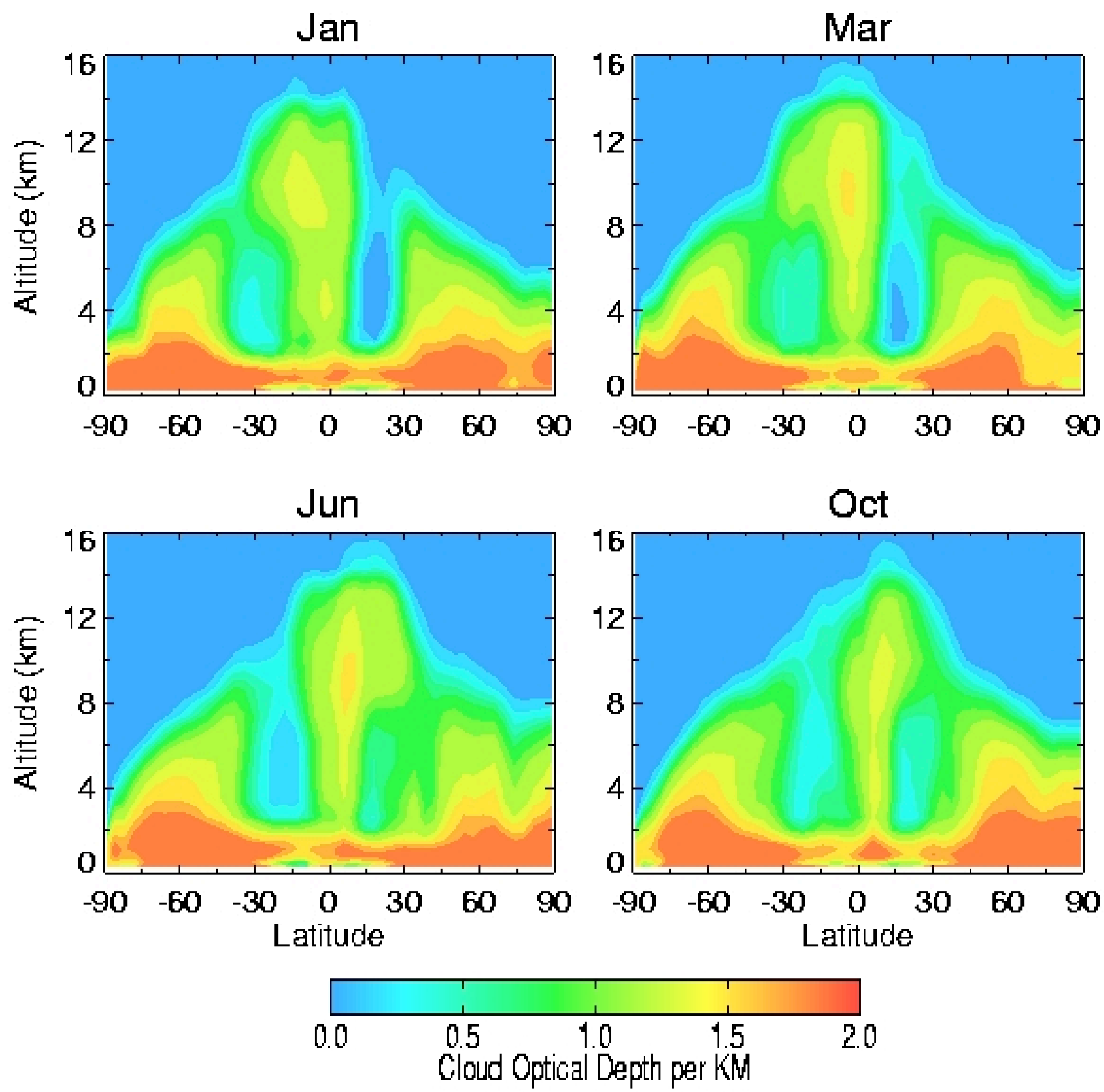
Cloud vertical distribution is more important than the magnitude of COD in terms of the radiative impact on <u>global</u> tropospheric chemistry!

Sensitivity of Global Mean OH to COD Magnitude (LIN, GEOS-3, 2001)

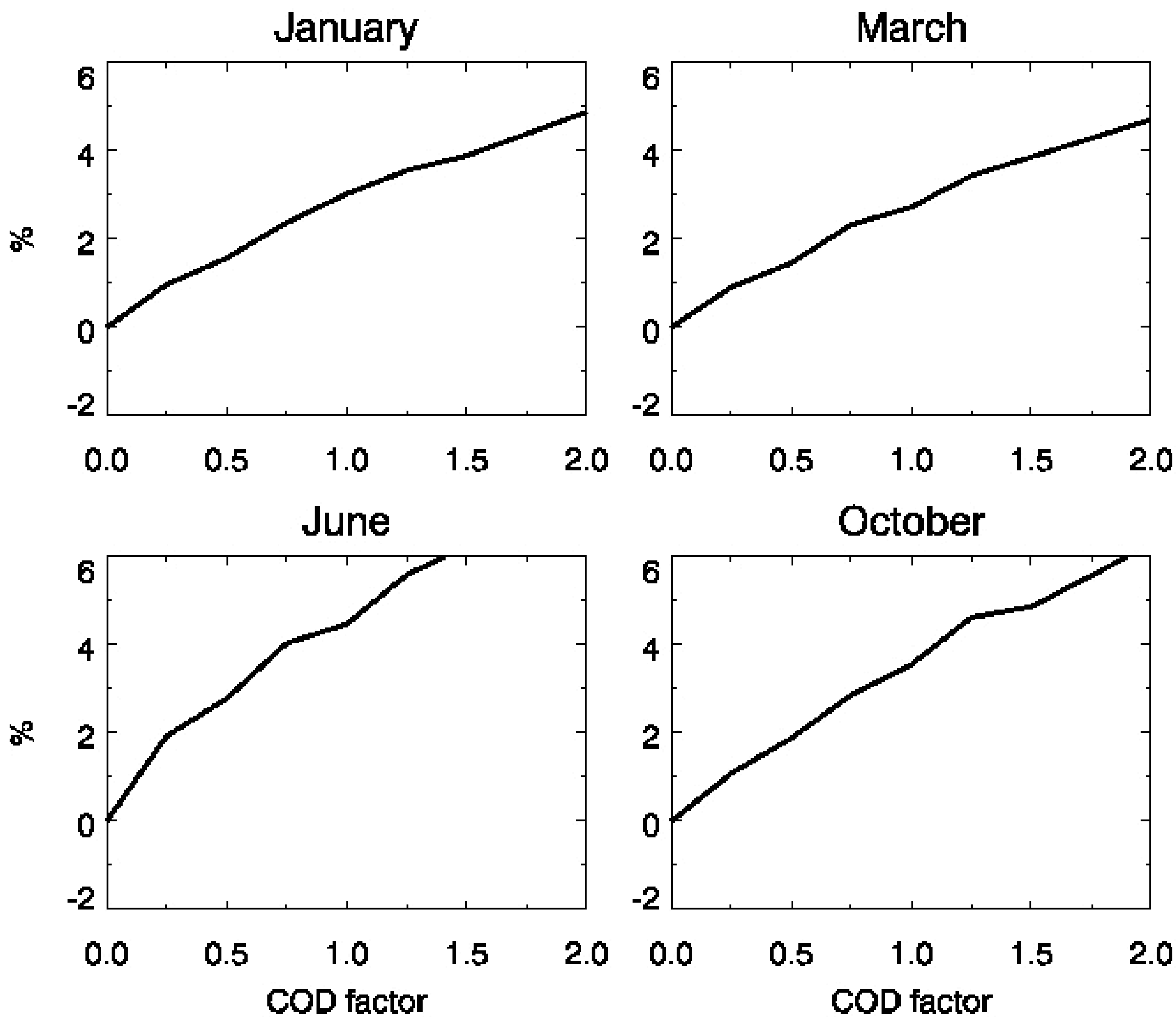


Global average effect is modest for all CODs.

Seasonal Variation of GEOS-3 COD



Sensitivity of Global Mean O_3 to COD Magnitude (LIN, GEOS-3, 2001)



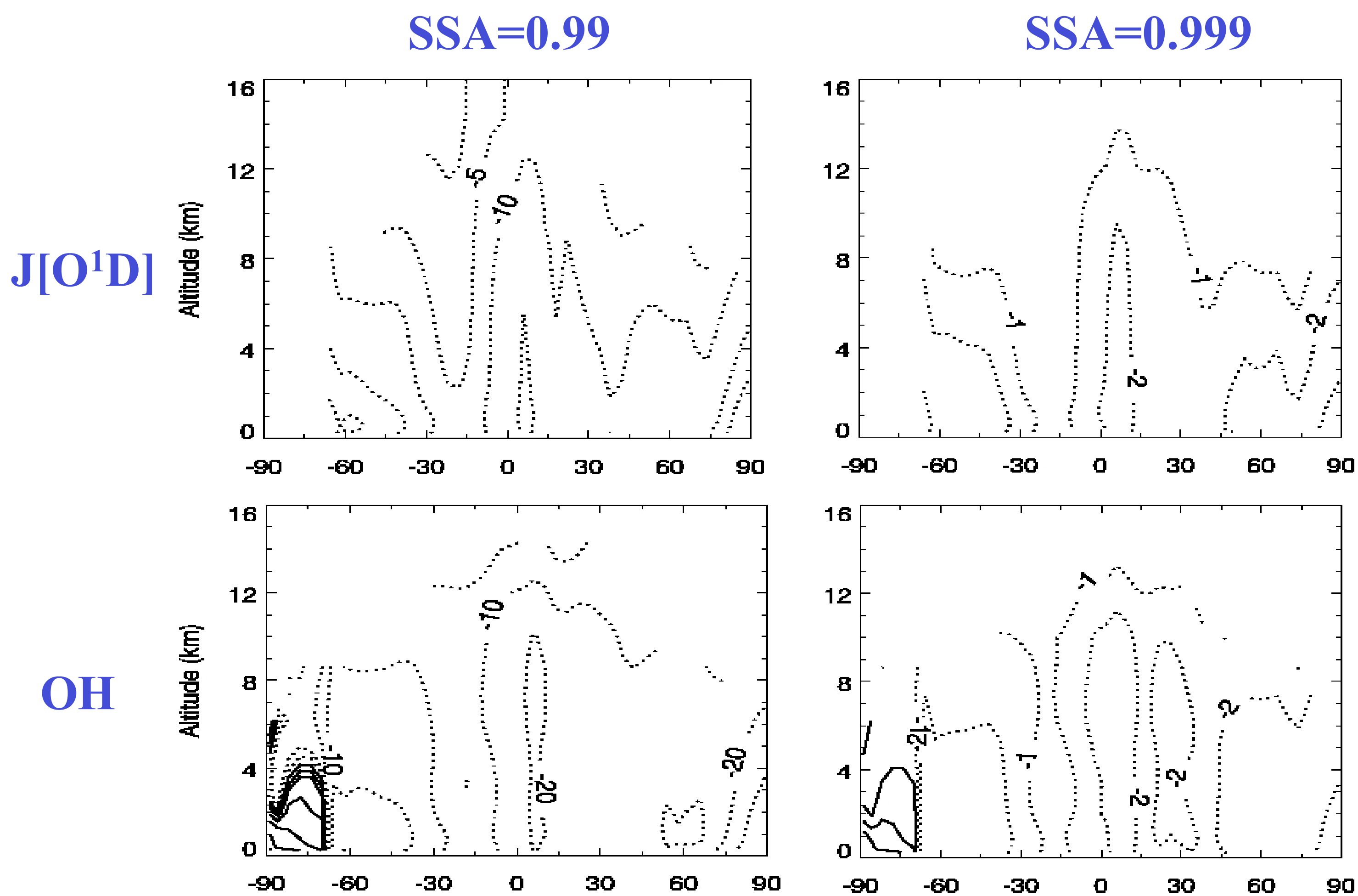
Cloud Single-Scattering Albedo (SSA)

- Pure water droplets: SSA=0.999990 - 0.999999 in the UV wavelength range [Hu and Stamnes, 1993]

- Clouds containing black carbon: SSA=0.999 – 0.9999 at 550 nm [Chylek et al., 1996]

- A cautionary note: SSA = 0.99 is NOT even close and is too low!

Sensitivity of J[O^{1}D] and OH to Cloud Absorption (% Changes Relative to SSA=1; GEOS-3, June)



Again, SSA=0.99 is NOT even close to SSA=1

Conclusions

- The dominant radiative effect of clouds is to influence the **vertical redistribution** of the intensity of photochemical activity while the global average effect remains modest. **This contrasts with previous studies**.

- Using the maximum-random overlap scheme or the random overlap scheme (vs. linear assumption) reduces the impact of clouds on photochemistry, but global average effect remains modest.

- The radiative effect of clouds in global CTMs is more sensitive to **cloud vertical distribution** than to the magnitude of COD.

- The radiative effect of clouds on trop chem and its associated uncertainties **need to be assessed within the GMI framework**.